UTILIZATION OF NEURAL NETWORKS FOR GPU-ACCELERATED DIFFUSION MONTE CARLO FOR VIBRATIONAL PROBLEMS

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In recent years, there has been increasing interest in combining machine learning with computational chemistry. Many machine learning applications in theoretical chemistry have focused on the development of potential energy surfaces (PES) based on *ab initio* data points[a]. Another computational resource that has gained popularity in recent years is the use of Graphics Processing Units (GPUs) to accelerate calculations such as molecular dynamics simulations [b]. In this work, we take advantage of the knowledge gained from both of these areas, and apply these approaches to the evaluations of the potential energy for Diffusion Monte Carlo (DMC) calculations. Diffusion Monte Carlo is a stochastic simulation approach through which an ensemble of localized functions (walkers) converges to the ground state vibrational wave function at long propagation times. This is accomplished through the ensemble randomly sampling a potential energy surface. These calculations require on the order of 10^8 to 10^10 potential energy evaluations, making the call to a potential energy surface the most expensive part of these calculations.

To this end, we have developed a general technique for "learning" already existing potential energy surfaces using neural networks, which can then easily be evaluated on GPUs using TensorFlow. The technique entails running a small-scale DMC simulation to collect training data, coordinates and energies, and using this data to fit a subspace of the potential energy surface that is relevant for the DMC simulations. We find that using the neural network potential energy surface maintains the fidelity of the wave function while also enabling the use of ensemble sizes orders of magnitude larger than previously used for the same amount of wall time. We test this method on small systems such as $CH5+$, $H2O$, and water dimer.

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[a] Manzhos, S.; Carrington, T. Chem. Rev. Article ASAP. https://doi.org/10.1021/acs.chemrev.0c00665

[b] Abraham, M. J.; Murtola, T.; Schulz, R.; Páll, S.; Smith, J. C.; Hess, B.; Lindahl, E. SoftwareX 2015, 1–2, 19–25